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A NEW MIDINFRARED LASER IN HYDROGEN

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Introduction: The basic research program for high energy lasers (HEL) at the U.S. Army Missile Command has had a major task in the area of new laser concepts for future Army HEL applications. A theoretical and experimental program to explore the possibility for developing a storage laser in hydrogen was carried out during FY 80-82 and resulted in the discovery of the laser reported here.

A storage laser is defined as a device which has the ability to store large amounts of energy (vibrational or electronic) without large collisional or radiative losses. There are a number of methods by which the stored energy can be extracted as stimulated emission. e.g., (i) by collisional transfer to another species with a large stimulated optical cross section, (ii) by an oscillator/amplifier cavity configuration (provided the saturation flux of the storage medium is not too large), or (iii) by temporarily increasing the stimulated cross section of the medium by such means as a field induced dipole.

Nitrogen is a prime example of a storage medium. Energy can be efficiently pumped into the vibrational mode of N_2 via discharge techniques: the collisional losses are small at room temperature and, of course, the radiative cross section for vibration/rotation transitions is zero; and nitrogen can efficiently transfer its vibrational energy to CO_2 . Therefore, the N_2/CO_2 laser is an example of type (i) storage laser.

Practical examples of type (ii) storage lasers are more difficult to find because they require a value of optical cross section that is large enough to permit a realizable saturable flux and low enough to prevent rapid loss of energy by spontaneous radiation. Optical cross sections of 10^{-19} - 10^{-20} cm^2 for visible transitions are required. Examples of this type of laser are the Group VI lasers - $O(^1S)$, $S(^1S)$, and $Se(^1S)$ - which were investigated as potential drivers for fusion reactors.

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The type (iii) storage lasers are, at present, only theoretical concepts. In such a laser the storage medium is pumped and, subsequently, a significant dipole moment is temporarily created in the molecule by impressing an electric field across the gas. Several investigators have proposed lasers of this type using radiation fields, as from a high intensity pulsed laser (1) or static external field from d.c. or low frequency a.c.(2). The problems in accomplishing this are formidable, and, to date, no demonstration of a field induced laser has been made.

Besides N_2 , the molecules H_2 and D_2 * are good potential storage media. The available electron impact data for H_2 indicate that it should be possible to pump into the vibrational mode of H_2 , D_2 and HD via an electric discharge with a controlled E/N (discharge field strength per gas density). The vibrational energy transfer kinetics of H_2 and D_2 have been experimentally measured and indicate that collisional losses at modest temperatures are small. In principle, the molecular system encompassing H_2 , D_2 , and HD can provide examples of all three types of storage lasers: For type (i) we have the H_2/HCl and D_2/HCl transfer electric discharge lasers (EDL), type (ii) is provided by HD itself since it has a small, permanent dipole moment, and for type (iii) H_2 , D_2 , and HD can all be considered for field induced dipole lasers.

Research on HD: Our work started with the analysis of HD as a type(ii), and H_2 as a type (iii) storage laser. First order calculations, in which kinetics processes were ignored, showed the conditions under which we could expect a sensible gain from either H_2 or HD based on the required partial population inversions. A major job for developing useable external fields would have been required for the H_2 laser, therefore, we elected to try the HD laser first.

Requirements for Laser Action in HD: The population distribution for threshold laser oscillation was calculated from a two level model in which the ground vibrational state, with population, N_l , was the lower state of the first vibrational state, with population, N_u , was the upper state. The required inversion density was obtained from the solution of the equations:

$$1. N_u - N_l(2J_u + 1)/(2J_l + 1) = \Delta N, \text{ and } 2. N_u + N_l = N$$

J_u and J_l are the rotational quantum numbers. The required inversion density is $\Delta N = g/s$ where g is the threshold small signal gain and s is the stimulated emission cross section.

* D is used in this text, as commonly done, to represent deuterium.

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The Einstein A coefficient was derived from data in (3); Doppler line broadening was assumed based on the physically realizable operating conditions (press = 4. torr and temp = 77°K to 300°K). Also thermal equilibrium was assumed among the rotational states. An estimate of the gain per centimeter required for threshold oscillation was made for state-of-the-art dielectric coated mirrors, $R_1=.995$ for the back mirror and $R_2=.98$ for the output mirror with no diffraction loss (implies a concave mirror), and a cavity length of 6.3 meters. A sensible output was assumed to exist when an absolute gain of 10^{16} was achieved, i.e., 10^{16} photons were produced in a coherent output. Threshold gain is:

$$3. \quad g = (2L)^{-1} \ln(R_1 \times R_2)^{-1} = (1260.)^{-1} \ln(.995 \times .98)^{-1} = 2.0 \times 10^{-5} \text{ cm}^{-1}.$$

The stimulated emission cross sections, s , were calculated and are: for 77°K and 300°K, respectively, for $P(1)$, $6.7 \times 10^{-22} \text{ cm}^2$ and 3.4×10^{-22} and for $P(2)$ $8.9 \times 10^{-22} \text{ cm}^2$ and $4.6 \times 10^{-22} \text{ cm}^2$. Thus the lowest, and presumably easiest to get, population inversion is for the $P(2)$ line at 77°K. In this case:

$$4. \quad N = g/s = 2.0 \times 10^{-5} \text{ cm}^{-1} / (8.9 \times 10^{-22} \text{ cm}^2) = 2.2 \times 10^{16} / \text{cm}^3.$$

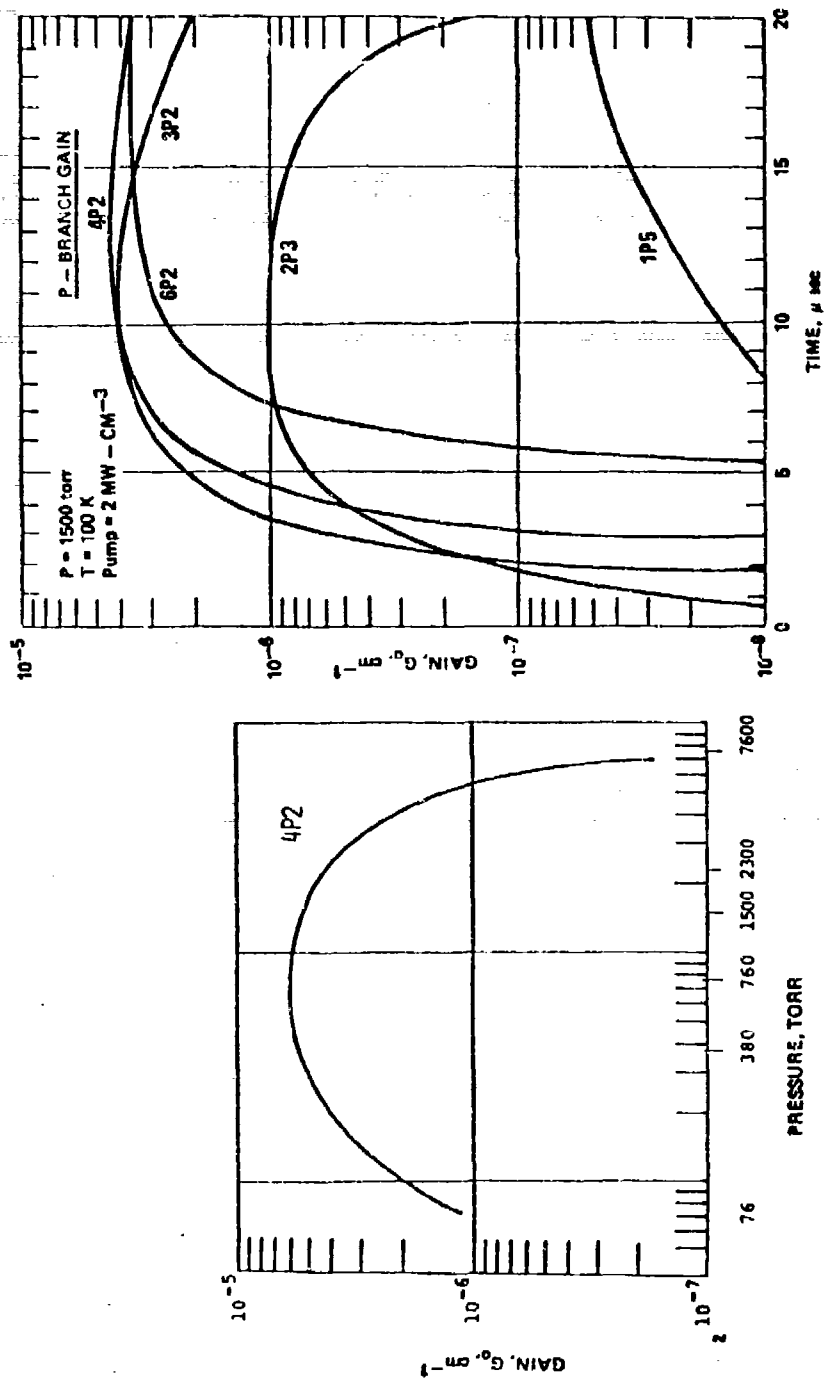
The total population at 4. torr and 77°K is $5 \times 10^{16} / \text{cm}^3$ and the fraction of the gas required to be in the upper state to meet the gain threshold is .04. We assumed, as a working hypothesis based on (4), that we could obtain sufficient population to overcome mirror losses plus about the same amount of gain to build up the laser flux. Under this gain the time to produce 10^{16} photons from one initiating photon is:

$$5. \quad t = \ln(N)/(c \times a) = \ln(10^{16} / (2 \times 10^{-5} \times 3 \times 10^{10})) = 60. \text{ microsec.},$$

where c is the velocity of light and a is the small signal gain.

The low gain requires that a long path, minimum loss laser be constructed to provide the best possible chance for laser action in HD. The laser described later in the text resulted from this requirement. It includes both a long path length, for maximum gain per "round trip," and a hemispherical cavity for minimum diffraction losses.

A detailed kinetics computer program was developed concurrently with the laser experiment. This program explicitly addressed the calculation of gain in the laser as a function of time for various electrical pumping conditions using a finite-rate kinetics analysis (5). The results indicate that small gains are obtainable. However, the magnitude and temporal duration of the gain are not sufficient to meet the gain-time requirements shown in equations, 3 and 5, above. Figure 1. shows the computed maximized gain occurring at 1500. torr (not 4. torr as in our experiment) and figure 2. shows that sufficient gain still falls short of the minimum time to produce a sensible output. With



Fig(1). Pressure Dependence of Gain for 4P2 Transition in HD

Fig(2). Small Signal Gain vs Time for HD

these computational results in hand and having not gotten laser output experimentally on HD we immediately turned our attention to making a transfer-storage laser, type (i).

The H₂/HCl and D₂/HCl Experiments: The laser was then converted, by change of mirrors, to run as a type (i) H₂/HCl or D₂/HCl laser. It was in this configuration that the presently reported lasers were first seen. The work on HD showed the sensitivity of the proposed experiments to gain so that working back through the time and threshold gain problem we found the conditions under which laser action could be detected in a few microseconds. The minimum gain for 1 microsec. generation of 10¹⁶ photons is:

$$6. \quad a = \ln(N)/cxt = 36.8/(3 \times 10^{10} \times 1 \times 10^{-6}) = 1.2 \times 10^{-3} \text{ cm}^{-1}$$

Previous work (6) showed that this gain might be achievable. Our experiments produced lasers in the midinfrared, first in H₂/HCl and later in D₂/HCl, but they were both found to be solely associated with H₂ and D₂. We stopped work on H₂/HCl and D₂/HCl to investigate these lasers. As will be explained later, they are newly discovered electronic transition lasers and not storage lasers.

Experiment Design: The experiment (7) is sketched in figure 3. The discharge tube consisted of a 2.5 cm diam Pyrex conical glass pipe. This included two, 3.0 m sections plus end Ts', discharge electrodes, mirror assemblies, and a center 5. cm x 30. cm cross section with an appended cold trap. The cavity output end used a flat sapphire window-mirror, dielectric coated to 95% reflectivity at 3.7 microns. A concave Pyrex mirror, 20 m radius of curvature, dielectric coated to 99+%, formed the other end. Both mirrors were partially transmitting in the visible, and a He-Ne laser was used for alignment of the cavity. Output was detected on a gold-doped germanium, liquid nitrogen-cooled detector. Detector voltage, discharge current, and discharge voltage were recorded on a storage oscilloscope.

The discharge was produced by a Marx bank fired through a resistor and across a gap into the laser tube. The bank consisted of four stages, each a 0.032 microfarads, 125kV, fast discharge capacitor. Normal charge voltage was in the range between 60 and 75 kV with corresponding erected voltages of 240 to 300 kV. Voltage was measured using a 10:1 RC voltage divider with a high voltage oscilloscope probe to provide an overall attenuation of 10000:1. Discharge current was measured by an inductive current transformer in the ground return circuit.

Typical operating conditions were: gas pressure, 2-8 Torr; current, 100-1200 amperes; run voltage (as opposed to initiation voltage) 10-50 kV; discharge duration, 0.5-8 microsec. Current, run voltage, and discharge times were primarily dependent on the discharge series resistors, which were wire wound, ceramic coated and ranged in value from 25-1000 ohms.

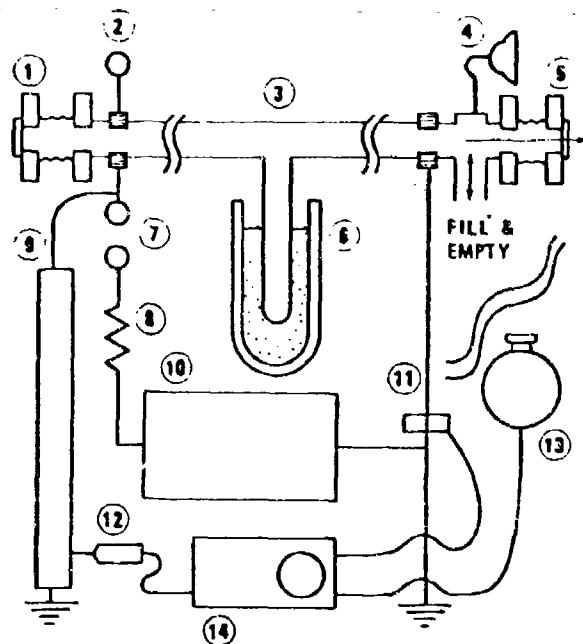


Fig (3). Experimental schematic for the laser: (1) concave mirror on adjustable sealed mount; (2) corona ring for high voltage end; (3) two 3. m sections of 2.5 cm diam Pyrex conical glass pipe; (4) pressure gauge, 0-20Torr; (5) output mirror on adjustable sealed mount; (6) liquid nitrogen cold trap; (7) discharge peaking gap; (8) current limiting resistor; (9) 10:1 voltage divider; (10) four-stage Marx bank; (11) current loop; (12) high voltage probe; (13) Au-Ge detector; and (14) wideband storage oscilloscope.

In the initial phase of the H_2 research, most experiments were done at room temperature; however, some were done with the tube in contact with a liquid nitrogen bath. This gave an estimated gas temperature of 100°K, inferred from pressure change in the tube during cooling. Some changes in laser action were noted during cold tests; however, these may have been caused by alignment problems and no conclusions were drawn from this experiment. The appended cold trap was also tested and after an hour of trapping at liquid nitrogen temperature no difference in laser performance was noted.

A quarter meter grating monochromator with a 150 line/mm grating and an 800 micron slit system was used for coarse location of the lines. The wavelength was measured using a half meter Ebert monochromator with a 148-lines/mm grating blazed at 5 microns. Slit widths between 400 and 100 microns were used, the former to scan for line center and the latter to obtain the wavelength. Calibration was by comparing dial settings to

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the 5th, 6th, and 7th order of the He-Ne laser. The wavelength measurements were not sufficiently accurate to include the fine correction for dispersion.

The H₂ Line at 3.767 Micron: - Parametric variation experiments were being performed on a mixture 95% hydrogen/5% hydrogen chloride when the laser was first discovered. Two distinct laser output patterns were noted. During the high current discharge a very short (0.5 microsec.) relatively high intensity (7 volt detector output) pulse close to the leading edge of the current pulse was observed. As the peak current was lowered, i.e., series resistance increased, the pulse occurred later in the current cycle, was lower in peak intensity, and stretched out to several microseconds. Eventually, when the 500 and 1000 ohms resistors were used, and the current was down to 150 amps or less, the pulse took on a distinctively different shape. It appeared after the current peak, followed the current shape for 3-4 microsec. and then terminated. Figure (4) shows oscilloscope traces for a short pulse; Figure (5) shows similar traces for a long pulse.

The question of the source of the laser transition was addressed by trying various mixes of gases, by the cold tube and cold trap experiments, and by removing spurious sources of unknown gases insofar as was possible. Mixtures of hydrogen-nitrogen, hydrogen-helium, and hydrogen-hydrogen chloride had varying effects on the output, but laser action continued until dilution effects dominated. However, a deuterium-hydrogen mixture appeared to suppress the laser action. No significant difference in output was noted between commercial grade and spectroscopic grade hydrogen. Laser action was terminated by cavity detuning; a tuned to detuned intensity ratio was at least 4 orders of magnitude. Particular attention was given to the possibility that the transition might be in hydrogen chloride, since the laser was first seen with a hydrogen-hydrogen chloride mixture in the tube, and only laser transitions known to us near this wavelength are hydrogen chloride and deuterium fluoride transitions. In hydrogen chloride the V(2-1)P(6) vibration-rotation transition lies at 3.77 microns. Spectrometric measurements, however, showed that the wavelength was definitely different from this hydrogen chloride line, and all the experiments mentioned here indicated that only hydrogen was involved in the laser.

Other Lines in H₂ and D₂: The D₂ lines were discovered under conditions similar to those of the H₂ line. In the case of D₂ a mixture of 95% deuterium/5% hydrogen chloride was electrically excited in the laser cavity. No laser action was observed at room temperature; however, because the kinetics would be more favorable at low temperature we cooled the laser with liquid nitrogen and repeated the experiments. Laser action was noted under these circumstances at 4.60 microns. Again, tests were made

to determine the source and all but deuterium was eliminated by the same procedure used on hydrogen.

Replacement of the mirrors was required when the coating came off the concave cavity mirror. The new mirrors were coated for the 3.7-4.7 micron band with maximum reflectivity on the concave mirror (99+%) and from 35% to 95% on the flat mirror (output end).

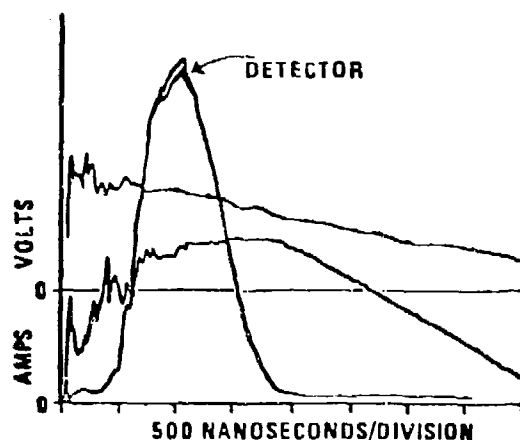


Fig (4). Short pulse, 50 ohm series resistance

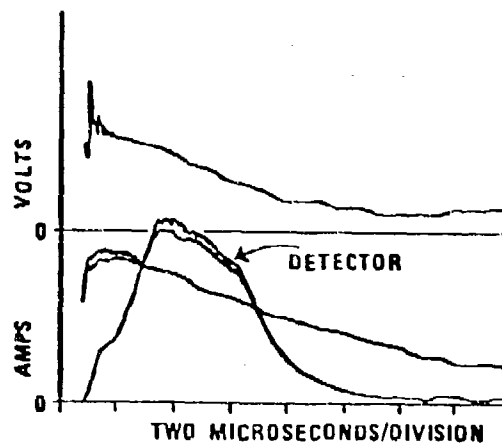


Fig (5). Long pulse, 500 ohm series resistance

All experiments were repeated with the new set of mirrors. Altogether three lines were found in hydrogen and three in deuterium. The hydrogen lines, 3.70 microns, 3.77 microns, and 3.84 microns were obtained at room temperature. The deuterium lines were found only when the laser was cooled with liquid nitrogen. The lines are, in order of decreasing intensity, 4.60 microns, 4.52 microns, and 4.70 microns. No laser lines were found in deuterium hydride at either temperature.

Spectroscopic Assignment of Lines: Following the work of (8), (9) and using the spectroscopic constants and notation of (10) we matched the 3.70 micron line to the singlet transition E-B, V(0-5) P(2) and the 3.77 micron line to the singlet transition K-C, V(1-6) P(4). A kinetically plausible set for all three lines was not found in this series.

Examination of the triplet structure produced entirely different results in which the experimental and calculated wavelengths for hydrogen are closely matched and fall into a kinetically linked pattern in the $a^3\Sigma_g^+ - c^3\Pi_u$ band as follows: 3.70 microns, V(3-2)P(2); 3.77 microns,

V(2-1)P(2); 3.84 microns, V(1-0)P(2). The set of calculated lines in this band which fall close to the ones observed in hydrogen are:

TABLE I. Calculated wavelengths in the $a^3\Sigma_g^+ - c^3\Pi_u$ band of H_2

	microns		microns		microns
V(1-0)P(1)	3.76	V(2-1)P(1)	3.69	V(3-2)P(1)	3.63
P(2)	3.84	P(2)	3.77	P(2)	3.71
P(3)	3.93	P(3)	3.85	P(3)	3.78
				P(4)	3.85

The temperature dependence of the rotational state population is given by:

$$7. J_{\max} = .59 (T/B)^{1/2} - .5$$

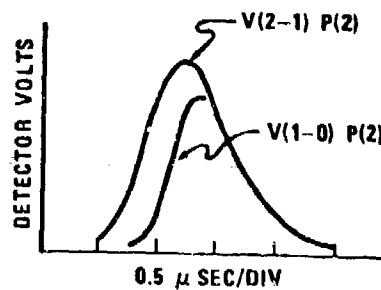
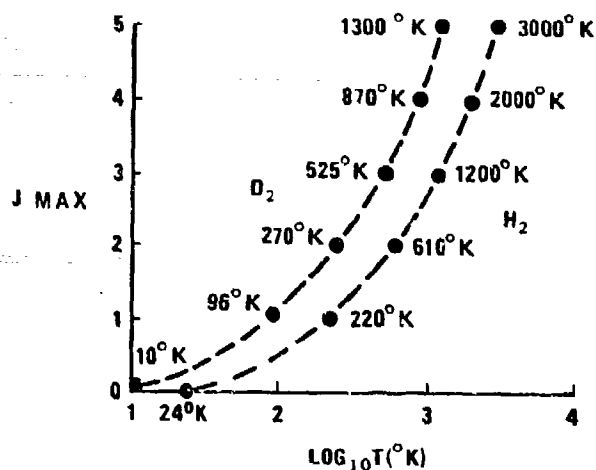
Figure 6. shows the graph of this function for hydrogen in the $a^3\Sigma_g^+$ state which $B = 34.\text{cm}^{-1}$ and for deuterium in the $c^3\Pi_u$ state in which $B = 15.\text{cm}^{-1}$. Reference (10) indicates that the upper laser level in D_2 may be $c^3\Pi_u$ state instead of the $a^3\Sigma_g^+$ state. P(2) appears to be the preferred rotational transition for both gases, which matches the experimental observation that hydrogen produces lasers at 3000K and 1000K. Experimentally, we find that the V(2-1) transition in H_2 , the strongest line, occurs first and is followed in about 0.2 microsec. by the other two lines. This is not unusual in cascading lasers; it is an indication that the laser action on the main line kinetically produces conditions for the other two lines to appear. Good computational modeling should show this effect, too. This delay is shown in the oscillograph traces of Figure 7.

An identical assignment cannot be made for deuterium: first, because its $c^3\Pi_u$ state, lies above the $a^3\Sigma_g^+$ state and second, because the vibrational and rotational coefficients (except B) are not known. In principle the laser lines discovered in deuterium could be used to produce spectroscopic coefficients. This will be tried at a later date, particularly if additional lines can be found and more accurate wavelengths determined.

Cavity Properties: The laser cavity composed of a plane output mirror and a concave back mirror, figure 8, may be considered to be half of a symmetric resonator. It has an effective mirror diameter of 2.5 cm and a full cavity length of 1260 cm (twice the mirror spacing). A typical wavelength is $4.\times 10^3$ cm so the Fresnel number is approximately 3. This configuration thus has negligible diffraction loss. The resonator g parameter is:

8. $g = g_1 = g_2 = 1 - L/R = 1 - 1260/2000 = .37$

and the stability factor, g_2 is 0.137, signifying a stable oscillator.



Fig(6). J max for H_2 , $a^3\Sigma_g^+$ and for D_2 , $c^3\Pi_u$

Fig(7). $V(1-0)P(2)$ line delay with respect to $V(2-1)P(2)$

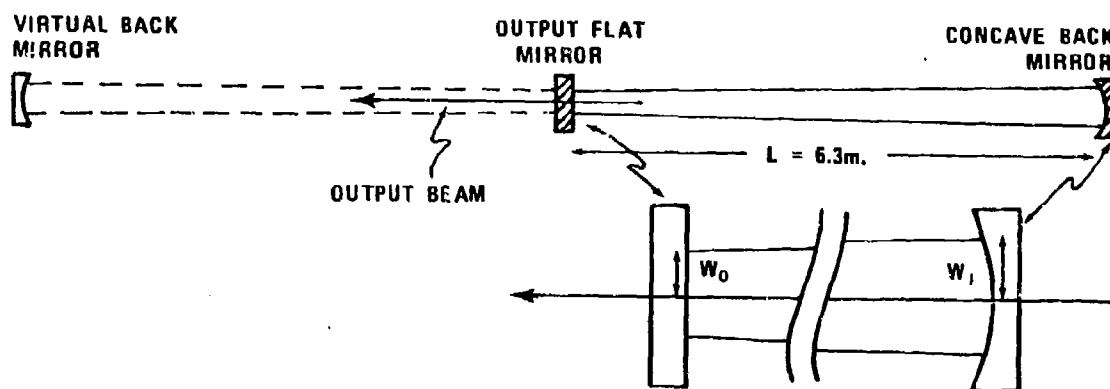


Fig. 8. Equivalent Cavity

The beam waist, radius w_0 , is at the flat output mirror and is,

9. $w_0 = (2L/\pi)^{1/2} (g^2(1-g^2))^{1/4} / (2g-2g^2)^{1/2} = .35 \text{ cm}$

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The back mirror spot size, w_1 , is

$$10. \quad w_1 = (2L/\pi)^{1/2} (1-g^2)^{-1/4} = .38 \text{ cm}$$

The average mode radius is .37 cm as compared to the discharge radius of 1.cm; thus the mode fills above .14 of the discharge volume.

Excitation Discharge Properties: The general circuit block diagram is shown in Figure 3. Properties of the discharge were calculated from the measured current and voltage and from previously calculated (4) transport properties of hydrogen and deuterium discharges. Energy distribution to vibration, rotation, dissociation and translation were determined as functions of the input parameters: pressure, temperature, voltage and current. Discharge properties are determined for hydrogen as follows: given current and voltage as functions of time as shown in Figure 9., we calculate the E/N values as functions of time. Drift velocities, W_d , for the electrons are then taken from the curves of W_d vs E/N (4). The free electron concentration and ion concentrations are calculated from these data by:

$$11. \quad n_e (=n_i) = I/(qxW_dxA)$$

where I is the measured current, q is the charge on the electron, and A is the cross section of the discharge. Likewise power input and the distribution of power to the various modes were tabulated as functions of time from the graphs of (6) which show energy distribution as obtained from a Boltzmann calculation.

The rate of electron-ion pair formation and the concentration of these are thus determined from measured and computed discharge properties. The properties associated with the discharge shown in Figure 9. are given in Table II below:

Table II, Properties of a Typical Discharge in Hydrogen

TIME (usec)	POTENTIAL (kV)	CURRENT (amps.)	POWER (kw/cm ³)	ENERGY (joules/cm ³)	EL.DENS. cm ⁻³	DIST. OF POWER		
						pd	pv	pl
0.5	110.	1200.	64.	.017	1.8×10^{14}	.70*	.18*	.07*
1.0	80.	1240.	48	.045	2.7×10^{14}	.51	.29	.13
1.5	52.	910.	23	.062	2.4×10^{14}	.32	.44	.20
2.0	35.	580.	9.8	.069	2.4×10^{14}	.05	.64	.27
2.5	25.	300.	3.6	.075	1.8×10^{14}	.0	.66	.31

*estimated by extrapolation

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Distribution of power terms represent the power at any time going in to the various uses as follows: P_d into dissociation, P_v into vibrational excitation, and P_T into thermal plus rotational excitation

On-set of laser action typically occurred, for the discharge with a 50 ohms series load, at 1. microsec. The gas properties at that time are noted in the Table II. Specific energy loading in the hydrogen at 1. microsec is 8500. joules/liter atmos.

This may be compared to the best loading for e-beam CO_2 lasers at one atmosphere which is about 900 joules/liter atmos. The eventual loading on the hydrogen in this example was 14,000. joules/liter atmos. at 2.5 microsec.

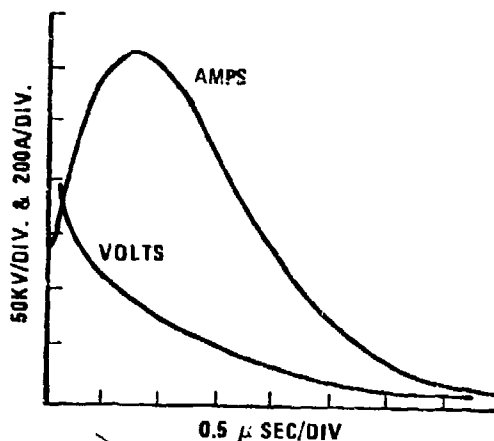


Fig. 9. Oscilloscope trace of current and voltage for the experimental laser (Fig. 3.) when the series load was 50 ohms.

CONCLUSION: The research program on hydrogen as a storage laser for high energy lasers applications has shown that HD certainly cannot be made into a storage laser under the operating conditions we had and perhaps cannot be so made under any circumstance. No conclusion was reached concerning H_2/HCl or D_2/HCl since serendipity produced a new set of lasers in the course of experiments on these gas mixtures. The new lasers, identified at least in hydrogen as being in the triplet states, may be the first step in successful demonstration of the hydrogen excimer (~~II~~).

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